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(54) Adhesive compositions.

(57) An adhesive composition comprising  
100 parts by weight of a block copolymer (a) containing  
of at least one polymer block composed mainly of a vinyl  
aromatic hydrocarbon and at least one polymer block com-  
posed mainly of butadiene in which

(i) the vinyl aromatic hydrocarbon content [S] is 10 to  
30% by weight,

(ii) the 1,2-vinyl content of butadiene portion [V] is 20 to  
50%, and

(iii) the relationship between said vinyl aromatic hydro-  
carbon content [S] and said 1,2-vinyl content of butadiene  
portion [V] satisfies the formula:

$$40 \leq [S] + [V] \leq 70$$

40 to 200 parts by weight of a tackifier (b), and, optionally  
contain  
various stabilizers; and a process for producing said  
composition.

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ADHESIVE COMPOSITIONS

1 FIELD OF THE INVENTION AND RELATED ART STATEMENT

FIELD OF THE INVENTION

The present invention relates to adhesive compositions which have excellent initial tack, adhesive  
5 strength and creep resistance and also possess an improved treatment capability at high-temperature. The present invention further provides compositions which are suitable for adhesive tapes, labels, etc.

RELATED ART STATEMENT

10 Adhesives have heretofore been used for various purposes. For example, adhesives have been used in adhesive tapes and labels. To coat an adhesive onto tapes, generally a method is employed which comprises dissolving the adhesive in a solvent to form an adhesive  
15 solution and coating a substrate with the adhesive solution by a means, such as roll coating or spray. However, the employment of such a solvent involves a multitude of problems from the standpoint of air pollution, fire, work environment and hygiene, etc., and  
20 in recent years, has come to be regarded as a serious social problem. As a means for eliminating such problems, it has recently been noticed that non-solvent type hot-melt adhesives have been used.

As base polymers of the hot-melt adhesives,

1 various polymers have been used, polymers such  
ethylene-vinyl acetate copolymers, block copolymers of  
conjugated dienes and vinyl aromatic hydrocarbons,  
ethylene- $\alpha$ -olefin copolymers, and polyester resins. In  
5 particular, block copolymers are preferred because of  
its relatively favorable balance between adhesive proper-  
ties and creep resistance. For example, Japanese Patent  
Publication Nos. 17037/69 and 41518/70 disclose adhesive  
compositions using a linear-chain block copolymer such as  
10 polystyrene-polybutadiene-polystyrene or polystyrene-  
polyisoprene-polystyrene. Japanese Patent Publication No.  
49958/81 discloses a hot-melt adhesive composition using a  
branched-chain block copolymer (polystyrene-polybutadi-  
ene)<sub>n</sub>X.

15 However, block copolymers of styrene and  
butadiene are insufficient in initial tack. Therefore, as  
base polymers for adhesive tapes and labels in which  
sufficient initial tack is particularly required, block  
copolymers of styrene and isoprene which have excellent  
20 initial tack, have recently been used often. But, in the  
case of adhesive compositions which comprises block  
copolymers of styrene and isoprene, when allowed to stand  
at high temperatures for a long period of time, resulted  
in cleavage of the molecular chain, the cleavage being  
25 caused by heat degradation. Consequently, the viscosity  
and the physical properties have been changed. Therefore,  
it is desirable to improve the initial tack of styrene-  
butadiene block copolymers which undergo no cleavage of

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1 the molecular weight. Various researches have been conducted on improving the initial tack of the  
styrene-butadiene block copolymers. For example, Japanese  
Patent Publication No. 20972/79 discloses a method using a  
5 hydrogenated aromatic petroleum resin as a tackifier. The  
tackifier had been prepared by hydrogenating a petroleum  
resin obtained by polymerization of a petroleum distillate  
comprising a polymerizable aromatic hydrocarbon as its  
main constituent. The ratio of hydrogenated aromatic ring  
10 in the aromatic component had been adjusted to 40 to 80%.  
Japanese Patent Application Kokai (Laid-Open) No. 2375/82  
discloses a method using a hydrogenated resin having a  
softening point of 50° to 160°C. The hydrogenated resin  
had been prepared by removing most of the vinylnorbornane,  
15 which was main product, by distillation thereof from a  
reaction mixture. The reaction mixture was produced by  
Diels-Alder reaction of 1,3-butadiene with cyclo-  
pentadiene. The thus obtained distillate having a  
dicyclopentadiene concentration of 10 to 70% and a  
20 concentration of tetrahydroindene and/or vinylcyclohexene  
of 30 to 80% was subjected to copolymerization by heating  
the distillate at 220° to 320°C, and hydrogenating the  
resin thus obtained. However, the initial tack is still  
not sufficiently improved even by such methods. Accord-  
25 ingly, there is a desire to seek further improvement in  
this area.

BAD ORIGINAL



1 OBJECT AND SUMMARY OF THE INVENTION

In consideration of such conditions, the present inventors have devoted themselves to research directed toward improving the initial tack of an adhesive composition containing a block copolymer of butadiene and vinyl aromatic hydrocarbon. Consequently, the present inventors have found that improved initial tack can be achieved by the use of a block copolymer of butadiene and vinyl aromatic hydrocarbon in which the vinyl aromatic hydrocarbon content of the block copolymer and its 1,2-vinyl content of butadiene portion are in specific ratios, which would specify the relationship between the two.

Further, the present inventors have found that the treatment capabilities at a high temperature can be further improved by incorporating into the composition comprising the above-mentioned specified block copolymer, a specific phenolic compound or the combination of a specific phenolic compound and a sulfur-containing stabilizer.

20 More particularly, the present invention relates to an adhesive composition comprising

100 parts by weight of a block copolymer (a) containing at least one polymer block composed mainly of a vinyl aromatic hydrocarbon and at least one polymer block composed mainly of butadiene in which

(i) the vinyl aromatic hydrocarbon content [S] is 10 to 30% by weight,

(ii) the 1,2-vinyl content of butadiene portion

1 [V] is 20 to 50%, and

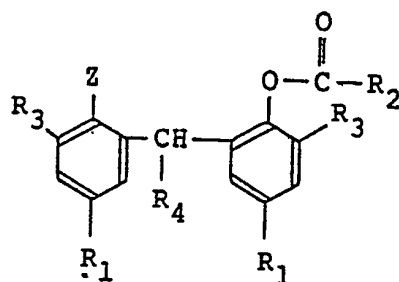
(iii) the relationship between said vinyl aromatic hydrocarbon content [S] and said 1,2-vinyl content of butadiene portion [V] satisfies the formula:

$$40 \leq [S] + [V] \leq 70$$

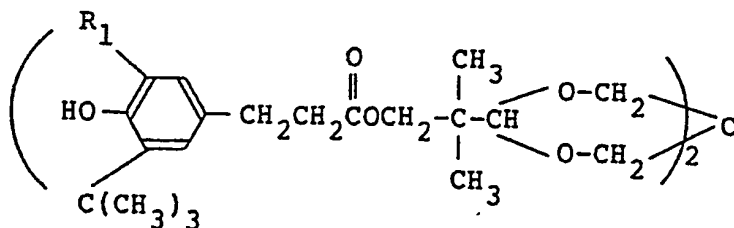
5 and

40 to 200 parts by weight of a tackifier (b).

Further, the present invention relates to an adhesive composition obtained by incorporating into the adhesive composition comprising the above-mentioned  
10 components (a) and (b), 0.05 to 5 parts by weight of at least one member of phenolic compounds (c) represented by the general formulae [I] and [II] selected from the group consisting of



[I] and,



[II]



10 an sulfur-containing stabilizer (d).

15 characterized with the stability of adhesive properties,

20 in parts by weight of each component per 100 parts by

25 "1,2-vinyl content of butadiene portion [V]" is given in a

- 1 butadiene units in said block copolymer (a) which corre-  
sponds to the total of butadiene units bounded in the form  
of both 1,4-structure and 1,2-vinyl structure therein.  
Said ratio can be calculated by determining 1,2-vinyl  
5 content in the butadiene portion using an infrared  
spectrometer, NMR apparatus or the like.

#### DETAILED DESCRIPTION OF PREFERRED EMBODIMENTS

- The block copolymer as component (a) used in the  
present invention contains at least one, preferably two or  
10 more polymer blocks composed mainly of a vinyl aromatic  
hydrocarbon and at least one polymer block composed mainly  
of butadiene. The term "polymer block composed mainly of  
a vinyl aromatic hydrocarbon" used herein means a  
copolymer block of a vinyl aromatic hydrocarbon and  
15 butadiene having a vinyl aromatic hydrocarbon content of  
50% by weight or more, and/or a vinyl aromatic hydrocarbon  
homopolymer block. The term "polymer block composed  
mainly of butadiene" used herein means a copolymer block  
of butadiene and a vinyl aromatic hydrocarbon having a  
20 butadiene content of more than 50% by weight, and/or a  
butadiene homopolymer block. The vinyl aromatic hydro-  
carbon in the copolymer blocks may be distributed either  
uniformly or taperingly. A plurality of portions wherein  
the vinyl aromatic hydrocarbon is uniformly distributed  
25 and/or a plurality of portions wherein the vinyl aromatic  
hydrocarbon is taperingly distributed may coexist in each  
block.



1           The vinyl aromatic hydrocarbon content [S] of  
the block copolymer as component (a) used in this inven-  
tion is 10 to 30% by weight, preferably 15 to 25% by  
weight, and the 1,2-vinyl content of butadiene portion [V]  
5 is 20 to 50%, preferably 25 to 45%. When the vinyl  
aromatic hydrocarbon content is less than 10% by weight,  
the creep resistance is low, and when it exceeds 30% by  
weight, the initial tack is insufficient. When the  
1,2-vinyl content of butadiene portion is outside the  
10 above range, the initial tack is insufficient. The block  
copolymer as component (a) used in this invention is  
preferably one in which the relationship between [S] and  
[V] satisfies the formula:

$$40 \leq [S] + [V] \leq 70$$

particularly preferably

$$45 \leq [S] + [V] \leq 60$$

15 When [S] + [V] is outside the above range, a composition  
being excellent in balance among initial tack, adhesive  
strength and creep resistance, cannot be obtained.

20           The vinyl aromatic hydrocarbon used in this  
invention includes styrene, o-methylstyrene, p-methyl-  
styrene, p-tert-butylstyrene, 1,3-dimethylstyrene,  
α-methylstyrene, vinyl naphthalene, vinylanthracene, etc.,  
with styrene being the most preferred. These compounds

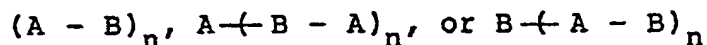
1 may be used either alone or as a mixture thereof.

As a process for producing the block copolymer used in the present invention, there are exemplified well-known processes. For example, such processes are  
5 disclosed in Japanese Patent Publication Nos. 19286/61, 24915/65, 17979/68, 31951/70, 32415/71, 14132/72, etc., but with the vinyl aromatic hydrocarbon content [S] and the 1,2-vinyl content of butadiene portion [V] being in the ranges specified for the present invention. The  
10 1,2-vinyl content can be adjusted by controlling the type and amount of a polar compound used as a vinyl modifier, the polymerization temperature, and the like in a polymerization method which comprises carrying out the polymerization in a hydrocarbon solvent by using an  
15 organolithium compound as the initiator.

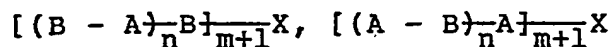
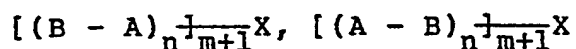
As the hydrocarbon solvent, there can be used, for example, aliphatic hydrocarbons, such as butane, pentane, hexane, isopentane, heptane, octane, isooctane, and the like; alicyclic hydrocarbons such as cyclopentane,  
20 methylcyclopentane, cyclohexane, methylcyclohexane, ethylcyclohexane, and the like; and aromatic hydrocarbons such as benzene, toluene, ethylbenzene, xylene, and the like. The organolithium catalyst includes, organomono-lithium compounds, organodilithium compounds, organo-  
25 polylithium compounds, etc. Specific examples of these compounds include, ethyl lithium, n-propyl lithium, isopropyl lithium, n-butyl lithium, sec-butyl lithium, tert-butyl lithium, hexamethylene dilithium, butadienyl

1 dilithium, isoprene dilithium, etc. The polar compound  
includes ethers, such as tetrahydrofuran, diethylene  
glycol dimethyl ether, diethylene glycol dibutyl ether and  
the like, amines such as triethylamine, tetramethylethy-  
5 lenediamine and the like, thioethers, phosphines,  
phosphoramides, alkylbenzenesulfonates, alkoxides of  
potassium and sodium, etc.

In the present invention, there can be used  
block copolymers whose polymer structures are represented  
.0 by the general formula:



(wherein A is a polymer block composed mainly of a vinyl  
aromatic hydrocarbon; B is a polymer block composed mainly  
of butadiene; the boundary between the blocks A and B need  
not necessarily be clear; and n is an integer of 1 or  
15 more), or by the general formulae:



(wherein A and B have the same meanings as defined above;  
X is a residue of a coupling agent such as silicon tetra-  
chloride, tin tetrachloride, epoxidized soybean oil or the  
like, or a residue of an initiator such as polyfunctional  
20 organolithium compounds or the like; and each of m and n

1 is an integer of 1 or more). Optionally, a mixture of  
block copolymers represented by the above general formulas  
may be used as component (a).

As the block copolymer used in this invention,  
5 those having a number average molecular weight of 10,000  
to 500,000 can be used, and those having a melt flow  
(temperature 200°C, load 5 kg) of 0.5 to 50 g/10 min,  
preferably 1 to 30 g/10 min are suitable. When the melt  
flow is less than 0.5 g/10 min, the viscosity of the  
10 resulting adhesive composition is increased, so that the  
coating operation on tapes and the like tends to be  
difficult. When the melt flow exceeds 50 g/10 min, the  
creep resistance tends to be poor.

When the adhesive composition of this invention  
15 is particularly required to have high creep resistance, it  
is preferable to use a block copolymer which comprises at  
least two polymer blocks composed mainly of a vinyl  
aromatic hydrocarbon and at least one polymer block com-  
posed mainly of butadiene and in which all the ends of the  
20 polymer chains are polymer blocks composed mainly of a  
vinyl aromatic hydrocarbon.

When the adhesive composition of this invention  
is particularly required to have high initial tack, it is  
preferable to use a block copolymer which comprises at  
25 least one polymer block composed mainly of a vinyl  
aromatic hydrocarbon and at least one polymer block  
composed mainly of butadiene and in which at least one end  
of the polymer chains is a polymer block composed mainly

1 of butadiene.

Further, when there is required a composition especially excellent in balance between creep resistance and initial tack, it is recommended to use a mixture of  
5 100 parts by weight of the former block copolymer which is excellent in creep resistance and 3 to 100 parts by weight, preferably 5 to 80 parts by weight of the latter block copolymer which is excellent in initial tack.

In the adhesive compositions of this invention,  
10 there can be used a block copolymer in which an atomic group containing polar-group is attached to at least one of the ends of the polymer chains of the block copolymer as component (a) specified in this invention. The creep resistance of the adhesive composition can be improved by  
15 attaching an atomic group containing polar group to the end of polymer chain. The term "atomic group containing polar group" used herein means an atomic group containing at least one atom selected from the group consisting of nitrogen, oxygen, silicon, phosphorus, sulfur and tin.  
20 Specific examples of the atomic group containing polar-group include atomic groups containing at least one polar group selected from the group consisting of carboxyl group, carbonyl group, thiocarbonyl group, acid halide groups, acid anhydride groups, carboxylic acid groups,  
25 thiocarboxylic acid groups, aldehyde groups, thioaldehyde groups, carboxylic acid ester groups, amide groups, sulfonic acid group, sulfonic acid ester groups, phosphoric acid group, phosphoric acid ester groups, amino group,

1 imino group, nitrile group, pyridyl group, quinoline  
group, epoxy group, thioepoxy group, sulfide group,  
isocyanate groups, isothiocyanate groups, silicon halide  
groups, alkoxy silicon groups, tin halide groups, alkyl  
5 tin groups, phenyl tin groups, etc. More specifically,  
there can be used, as component (a), terminally modified  
vinyl aromatic hydrocarbon-butadiene block copolymers  
having a vinyl aromatic hydrocarbon content and a 1,2-vinyl  
content of butadiene portion in the ranges specified in  
10 this invention, among the terminally modified block  
copolymers described in Japanese Patent Application No.  
224806/85. The tackifier as component (b) used in this  
invention is one which has heretofore been used as  
tackifier in hot-melt adhesives and the like, and it  
15 includes, for example, coumarone-indene resin, phenolic  
resins, p-tert-butylphenol-acetylene resins, phenol-  
formaldehyde resins, terpene-phenol resins, polyterpene  
resins, xylene-formaldehyde resins, synthetic polyterpene  
resins, aromatic hydrocarbon resins, aliphatic cyclic  
20 hydrocarbon resins, oligomers of monoolefins and  
diolefins, hydrogenated hydrocarbon resins, hydrocarbon  
resins, polybutene, polyhydric alcohol esters of rosin,  
hydrogenated rosin, hydrogenated wood rosin, esters of  
hydrogenated rosin and monohydric alcohols or polyhydric  
25 alcohols, turpentine series tackifier. More specifical-  
ly, there can be used the tackifier described in "Rubber  
and Plastic Compounding Agents edited by Rubber Digest  
Co., Ltd.". Particularly suitable tackifiers are terpene

1 resins, aromatic compound-modified terpene resins,  
alicyclic saturated petroleum resins, rosin esters,  
disproportionated rosin esters, completely hydrogenated  
rosin esters, aliphatic petroleum resins ( $C_5$ -aliphatic  
5 petroleum resins,  $C_5$ ,  $C_9$ -aliphatic/aromatic petroleum  
resins, etc.),  $C_9$ -aromatic petroleum resins and modified  
aliphatic petroleum resins, and these tackifier give  
compositions good in initial tack.

In this invention, the tackifier as component  
10 (b) is used in an amount of 40 to 200 parts by weight,  
preferably 60 to 150 parts by weight per 100 parts by  
weight of the block copolymer as component (a). When the  
blending amount of the tackifier is outside the above  
range, the adhesive properties of adhesive tapes using the  
15 resulting adhesive compositions are unsatisfactory.  
Therefore, it is not desirable.

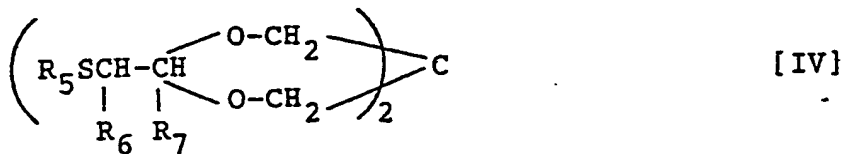
The adhesive compositions of this invention may  
be incorporated with phenolic stabilizers, sulfur-contain-  
ing stabilizers, phosphorous-containing stabilizers, amine  
20 series stabilizers, etc. in order to impart thereto  
thermal stability at high temperatures. Particularly  
preferable stabilizers include the phenolic compounds of  
the above general formulas [I] or [II] (component (c)) and  
combinations of said component (c) and the sulfur-contain-  
25 ing stabilizer (component (d)). In the phenolic compounds  
of the above general formulas [I] or [II], specific  
examples of the substituent  $R_1$  include methyl group,  
ethyl group, propyl group, isopropyl group, butyl group,

1 isobutyl group and tert-butyl group, though methyl group,  
isopropyl group or tert-butyl group is particularly  
preferred. The substituent  $R_2$  is an alkenyl group  
having 2 to 4 carbon atoms and specific examples thereof  
5 include ethenyl group, isopropenyl group, propenyl group,  
isobutenyl group, butenyl group, etc. Among them, ethenyl  
group is particularly preferred. The substituent  $R_3$  is  
preferably a tert-butyl group. Specific examples of the  
substituent  $R_4$  include hydrogen atom, methyl group, ethyl  
10 group, propyl group and butyl group, though hydrogen atom  
and methyl group are particularly preferred. The phenolic  
compound as component (c) is used in an amount of 0.05 to  
5 parts by weight, preferably 0.1 to 2 parts by weight per  
100 parts by weight of the block copolymer as component  
15 (a). When the using amount of component (c) is less than  
0.05 parts by weight, no improving effect on the treatment  
capabilities at a high temperature is brought about. On  
the other hand, even when it exceeds 5 parts by weight,  
the effect obtained is not larger than when the amount  
20 used is in the range specified in the present invention.

As the sulfur-containing stabilizer as component  
(d) used in the present invention, stabilizers commonly  
called "sulfur-containing stabilizers" among various  
stabilizers can be used. Specific examples thereof  
25 include dilauryl-3,3'-thiodipropionic acid esters,  
dimyristyl-3,3'-thiodipropionic acid esters, distearyl-  
3,3'-thiodipropionic acid esters, laurylstearyl-3,3'-  
thiodipropionic acid esters, ditridecyl-3,3'-thiodipro-



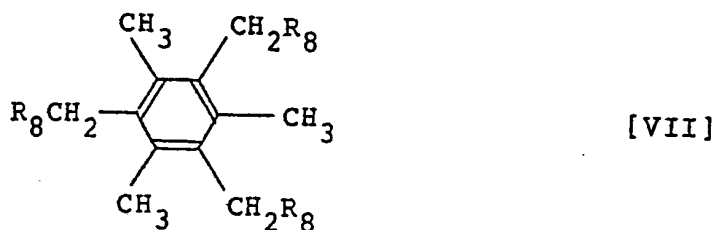
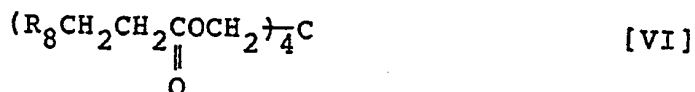
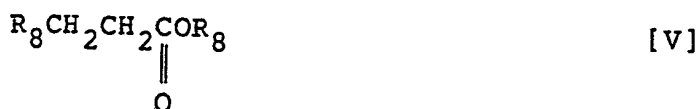
1 pionic acid ester compounds of the general formula [III]  
and [IV]:

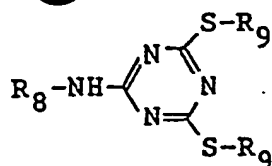


(wherein  $R_5$  is an alkyl group having 3 to 20 carbon atoms, specifically a hexyl group, an octyl group, a dodecyl group, an octadecyl group or the like, and is particularly preferably a dodecyl group; and each of  $R_6$  and  $R_7$  is a hydrogen atom or an alkyl group having 1 to 18 carbon atoms and is particularly preferably a hydrogen atom, a methyl group, an ethyl group, a propyl group or a butyl group), etc. Specific examples of the compounds of the general formula [III] or [IV] include pentaerythritol-tetrakis-(laurylthiopropionate), 3,9-bis(2-dodecylthioethyl)-2,4,8,10-tetraoxaspiro[5.5]undecane, and 3,9-bis(2-octadecylthiopropyl)-2,4,8,10-tetraoxaspiro[5.5]-undecane. The sulfur-containing compound as component (d) is used in an amount of 0.05 to 5 parts by weight, preferably 0.1 to 2 parts by weight per 100 parts by weight of the block copolymer as component (a). When the using amount of component (d) is less than 0.05 parts by weight, no improving effect on the treatment capabilities at a high temperature is brought about. On the other

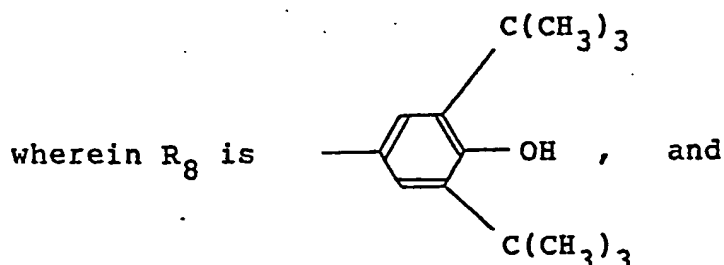
1 hand, over when it exceeds 5 parts by weight, the effect  
 obtained is not larger than when the using amount is in  
 the range specified in this invention. In the present  
 invention, compounds selected from the compounds of the  
 5 above general formulas [III] and [IV] are particularly  
 preferred as the sulfur-containing compound.

In the present invention, in order to improve  
 the treatment capabilities at a high temperature, at least  
 one phenolic compound selected from the group consisting  
 10 of the compounds of the general formulae [V], [VI], [VII]  
 and [VIII] shown below can be used as component (e) in an  
 amount of 0.05 to 5 parts by weight, preferably 0.1 to 2  
 parts by weight per 100 parts by weight of the block  
 copolymer as component (a):





[VIII]



- 1  $R_9$  is an alkyl group having 2 to 22 carbon atoms.

In the above general formulas, the substituent  $R_9$  is an alkyl group having 2 to 22 carbon atoms, preferably 4 to 20 carbon atoms. In particular, the substituent  $R_9$  is preferably an octadecyl group in the general formula [V] and an octyl group in the general formula [VIII].

In the present invention, if necessary, a phosphorus-containing stabilizer may be further incorporated in an amount of 0.05 to 5 parts by weight, preferably 0.1 to 2 parts by weight per 100 parts by weight of the block copolymer. Specific examples of the phosphorus-containing stabilizer include tris(nonylphenyl)phosphite, cyclic neopentane-tetraylbis(octadecylphosphite), tris(2,4-di-tert-butylphenyl)phosphite, 4,4'-butylidene-bis(3-methyl-6-tert-butylphenyl-di-tridecyl)phosphite, tetrakis(2,4-di-tert-butylphenyl) 4,4'-biphenylenediphosphate, cyclic

1 neopentane-tetraylbis(2,4-di-tert-butylphenyl)phosphite,  
etc. A composition good in color tone can be obtained by  
incorporating such phosphorus-containing stabilizers.

As the amine series stabilizers, there can be  
5 exemplified, for example, diarylamine type stabilizers  
such as diphenylamine, N-phenyl- $\alpha$ -naphthylamine, N-phenyl- $\beta$ -naphthylamine, butylated diphenylamine, styrenated  
diphenylamine, octylated diphenylamine, nonylated  
diphenylamine, and the like; alkylarylamine type stabi-  
10 lizers such as N-cyclohexylaniline, N-methylaniline,  
N-isopropylaniline, N-1,3-dimethylbutylaniline, N-(2-ethylhexyl)aniline, and the like; N,N'-diaryl-p-phenylenediamine type stabilizers such as N,N'-diphenyl-p-phenylenediamine, N,N'-diorthotolyl-p-phenylenediamine,  
15 N,N'-bis(2,4-xylyl)-p-phenylenediamine, N,N'-bistolyl-p-phenylenediamine, N,N'-ditolyl-p-phenylenediamine, N-tolyl-N'-xylyl-p-phenylenediamine, N,N'-dixylyl-p-phenylenediamine, N,N'-bis- $\beta$ -naphthyl-p-phenylenediamine, and the like; N-aryl-p-phenylenediamine type stabilizers  
20 such as N-phenyl-p-phenylenediamine, N-tolyl-p-phenylene-diamine, N-xylyl-p-phenylenediamine, and the like; N-aryl-N'-alkyl-p-phenylenediamine type stabilizers such as N-phenyl-N'-isopropyl-p-phenylenediamine, N-phenyl-N'-isobutyl-p-phenylenediamine, N-phenyl-N'-(1,3-di-  
25 methylbutyl)-p-phenylenediamine, N-phenyl-N'-cyclohexyl-p-phenylenediamine, N-phenyl-N'-octyl-p-phenylenediamine, and the like; N,N'-dialkyl-p-phenylenediamine type stabilizers such as N,N'-bis(1-methylheptyl)-p-phenylene-

1 diamine, N,N'-bis(1,4-dimethylpentyl)-p-phenylenediamine,  
N,N'-bis(1-ethyl-3-methylpentyl)-p-phenylenediamine, and  
the like; quinoline derivative type stabilizers such as  
2,2,4-trimethyl-1,2-dihydroquinoline, 2,2,4-trimethyl-1,2-  
5 dihydroquinoline polymers, 6-ethoxy-2,2,4-trimethyl-1,2-  
dihydroquinoline, and the like; and condensation product  
type stabilizers such as dehydrating-condensation products  
of aldol- $\alpha$ -naphthylamine or diphenylamine and acetone, and  
the like. These stabilizers may be used alone or in  
10 proper combination of two or more of them.

In the present invention, if necessary, a  
softener (component (f)) may be used. The softener as  
component (f) includes petroleum softeners, paraffin,  
fatty oil softeners, thermoplasticizers, etc. Specifi-  
15 cally, there can be used the softeners described in  
"Rubber and Plastic Compounding Agents" above. The  
softener as component (f) is used usually in an amount of  
150 parts by weight or less, preferably 5 to 100 parts by  
weight per 100 parts by weight of the block copolymer as  
20 component (a). When the using amount exceeds 150 parts by  
weight, the creep resistance of adhesive tapes using the  
resulting adhesive composition is low. Therefore, it is  
not desirable.

In the present invention, the creep resistance  
25 and the hardness can be improved by incorporating, as  
reinforcing resin, polystyrenes, polyethylenes, polypro-  
pylenes, ethylene-propylene copolymers, ethylene-butene  
copolymers, ethylene-vinyl acetate copolymers, or

1 thermoplastic resins such as relatively low-molecular-  
weight thermoplastic polyester resins, polyamide resins,  
polyphenylene ether resins and the like in an amount of 50  
parts by weight or less, preferably 2 to 40 parts by  
5 weight, more preferably 5 to 30 parts by weight per 100  
parts by weight of the block copolymer as component (a).  
When the blending amount of the reinforcing resin exceeds  
50 parts by weight, the initial tack is lowered. There-  
fore, it is not desirable. The adhesive compositions of  
10 the present invention may be used in admixture with other  
ordinary elastomer, for example, natural rubber, synthetic  
polyisoprene rubber, polybutadiene rubber, styrene-butadi-  
ene rubber, chloroprene rubber, ethylene-propylene rubber,  
acrylic rubber, polyisopreneisobutylene rubber, polycyclo-  
15 pentene (polypentenamer), vinyl aromatic hydrocarbon-  
conjugated diene block copolymers other than those  
specified in the present invention, etc., unless said  
adhesive compositions lose characteristics as adhesives.

#### EFFECTS OF THE INVENTION

20 Since the adhesive compositions of the present  
invention are excellent in initial tack, adhesive  
properties and creep resistance, they can be utilized in  
various adhesive tapes and labels, pressure-sensitive thin  
plates, pressure-sensitive sheets, various back pastes for  
25 fixing light-weight plastics molding, back pastes for  
fixing carpets, back pastes for fixing tiles, etc., and  
are effective particularly for use as adhesive tapes and

1 labels. Furthermore, the adhesive compositions of the  
present invention can be used as adhesives for plastics,  
rubber-like materials, foams, metals, wood, paper  
products, etc.

## 5 EXAMPLES

Examples are shown below not as limits on the  
scope of this invention but as representatives of this  
invention.

Examples 1 to 3 and Comparative Examples 1 to 7

10 Hot-melt adhesive compositions were produced by  
blending 100 parts by weight of each styrene-butadiene  
block copolymer having an A-B-A structure listed in Table  
1 (whose molecular weight has been adjusted so as to  
adjust the melt flow to about 5 g/10 min) with 100 parts  
15 by weight of an aliphatic petroleum resin (Quintone U-185,  
mfd. by Nippon Zeon Co., Ltd.) as tackifier, 30 parts by  
weight of a naphthenic process oil (Sonic Process Oil  
R-200, mfd. by Kyodo Petroleum Co., Ltd.), and 1 part by  
weight of 2,2'-methylenebis(6-tert-butyl-4-methylphenol)  
20 monoacrylate (hereinafter referred to as AO-1). In  
Comparative Example 7, a styrene-isoprene block copolymer  
was used as block copolymer.

Kraft adhesive tapes were produced by coating  
each of thus produced adhesive compositions on Kraft paper  
25 subjected to back treatment, and were subjected to the  
following performance assessments. The initial tack was

1 evaluated in the following manner according to J. Dow  
method [Proc. Inst. Rub. Ind., 1. 105 (1954)]. Each  
adhesive tape of 10 cm in length was attached to a slope  
on a stainless-steel plate inclining at an angle of 30°,  
5 after which stainless-steel balls of 32 sizes in the  
diameter range of 1/3 inch to 1 inch were individually  
rolled down the slope at an initial speed of zero from a  
position on the slope 10 cm above the upper end of the  
tape, and the initial tack was expressed in terms of the  
10 size of a ball having the largest diameter among balls  
which stopped on the adhesive tape. The adhesive strength  
was measured according to JIS Z-1522 by attaching each  
adhesive tape having a width of 25 mm and a length of 100  
mm to a stainless-steel plate, and peeling off the same at  
15 an angle of 180° at a speed of 300 mm/min at 23°C. The  
creep resistance was evaluated according to JIS Z-1524 by  
attaching each adhesive tape to a stainless-steel plate so  
as to adjust the contacted area to 25 mm x 25 mm, applying  
thereto a load of 1 kg at 60°C, and measuring the time  
20 required for the adhesive tape to be slipped down.

The results obtained are shown in Table 1 and  
indicate that the adhesive compositions in the ranges  
specified in this invention have satisfactory initial  
tack, adhesive strength, and creep resistance.



Table 1

	Block copolymer used			Ball tack (No.)	Adhesive strength (g/cm)	Creep resistance (min.)
	Styrene content [S] (wt.%)	1,2-Vinyl content [V] (%)	[S]+[V]			
Example	1	20	33	53	21	800
	2	25	30	55	19	760
	3	17	40	57	20	880
Comparative Example	1	20	11	33	16	740
	2	20	60	80	<3	610
	3	5	33	38	24	1020
	4	40	33	73	8	760
	5	30	50	80	7	730
	6	10	20	30	19	930
	7	SIS	(Note 1)			21
						700
						56

Note 1: styrene-isoprene block copolymer (K-1107, mfd. by Shell Chemical Co.)

1 Examples 4 to 14 and Comparative Examples 8 to 11

Adhesive compositions are produced in the same manner as in Example 1, except that each of the stabilizers listed in Table 2 and Table 3 was incorporated in place of AO-1, and their adhesive properties and melt viscosity after heating in air at 180°C for 48°C were measured.

As is evident from Table 2 and Table 3, the adhesive compositions of this invention showed satisfactory adhesive properties even after heating at a high temperature for a long period of time, and had such a low melt viscosity that they had excellent processability when used for producing an adhesive tape or label.

When the adhesive composition of Comparative Example 7 was heated in air at 180°C for 48 hours, after which its melt viscosity and adhesive properties were measured, it showed a ball tack of 23, an adhesive strength of 530 g/cm and a creep resistance of less than 20 minutes. Thus, the adhesive strength and the creep resistance were lowered. Moreover, the melt viscosity was as low as 2,000 poise as compared with the viscosity before the heating of 7,000 poise. Thus, the adhesive composition of Comparative Example 7 underwent a marked viscosity change and hence was poor in stability of processability.

Table 2

	Example							Comparative Example	
	4	5	6	7	8	8	9		
Blending amount of stabilizer (Note 2)	AO-1	1	1	0.2	2				
	AO-2		0.5	0.5		0.5			
	AO-3			0.25		0.2			
	AO-4			0.25					
	AO-5				0.5				
	AO-6	0.5	0.5		0.5	0.5	0.5		
	AO-7			0.5		0.5			
	AO-8					1	1		
	AO-9						0.5		
Melt viscosity (Note 3) (poise)	8500	6400	7100	7900	6000	>10000	>10000		

- cont'd -

Table 2 (cont'd)

Ball tack (No.)	17	19	19	18	21	10	13
Adhesive strength (g/cm)	710	750	740	740	780	680	700
Creep resistance (min)	130	140	150	135	160	105	110

Table 3

	Example								Comparative Example	
	9	10	11	12	13	14	10	11		
AO-1	0.5				0.2	2				
AO-10					1.0	0.2				
AO-11		0.5								
AO-12			0.5							
AO-13				0.5				0.01		
AO-14	0.5	0.5	0.5	0.5			0.01	0.5		
AO-9						0.2				
AO-6			0.5							
AO-7					0.5					
AO-8							1.0			
AO-2			0.5							
AO-4					0.5					
AO-5				0.5						

Blending  
amount of  
phenolic  
compound,  
etc.  
(Note 2)

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Table 3 (cont'd)

Melt viscosity (Note 3) (poise)	7500	8400	8000	9000	8100	6100	>10000	>10000
Ball tack (No.)	18	17	18	16	18	21	A large amount of a gel-like sub- stance was pro- duced, so that the adhesive proper- ties could not be evaluated.	
Adhesive strength (g/cm)	730	710	735	700	730	750		
Creep resistance (min)	140	130	135	120	130	160		

1 Note 2: There is shown a blending amount (parts by weight)  
per 100 parts by weight of the block copolymer.

The following compounds were used as the  
stabilizers.

5 AO-2: n-octadecyl-3-(3',5'-di-tert-butyl-4'-hydroxy-  
phenyl) propionate

AO-3: tetrakis-[methylene-3-(3',5'-di-tert-butyl-4'-  
hydroxyphenyl) propionate]methane

10 AO-4: 1,3,5-trimethyl-2,4,6-tris(3,5-di-tert-butyl-4-  
hydroxybenzyl)benzene

AO-5: 2,4-bis(n-octylthio)-6-(4-hydroxy-3,5-di-tert-  
butylanilino)-1,3,5-triazine

AO-6: tris(nonylphenyl)phosphite

AO-7: tris(2,4-di-tert-butylphenyl)phosphate

15 AO-8: 2,6-di-tert-butyl-4-methylphenol

AO-9: distearyl 3,3'-thiodipropionate

20 AO-10: a sulfur-containing stabilizer of the above  
general formula [IV] in which  $R_5$  is a  
dodecyl group and each of  $R_6$  and  $R_7$  is a  
hydrogen atom.

AO-11: a phenolic compound of the above general  
formula [I] in which Z is OH, each of  $R_1$  and  
 $R_3$  is a tert-butyl group,  $R_2$  is an ethenyl  
group, and  $R_4$  is a methyl group.

25 AO-12: a phenolic compound of the above general

O  
||

formula [I] in which Z is  $\text{OCR}_2$ ,  $R_1$  is a  
methyl group,  $R_2$  is an ethenyl group,  $R_3$  is

1           a tert-butyl group, and  $R_4$  is a hydrogen  
          atom.

AO-13: a phenolic compound of the above general  
          formula [III] in which  $R_1$  is a methyl group.

5           AO-14: a sulfur-containing compound of the above  
          general formula [III] in which  $R_5$  is a  
          dodecyl group.

Note 3: measured at 180°C by means of a Brookfield type  
          viscometer.

10   Examples 15 to 17

          Adhesive compositions were produced in the same  
manner as in Examples 1 to 3, except that 1.0 parts by  
weight of AO-1 and 0.5 part by weight of AO-10 were used  
as stabilizers. Their performance characteristics are  
15   tabulated in Table 4.



Table 4

	Block copolymer used			Ball tack (No.)	Adhesive strength (g/cm)	Creep resistance (min.)
	Styrene content [S] (wt. %)	1,2-Vinyl content (%)	[S] + [V]			
Example	15	20	33	53	20	165
	16	25	30	55	19	220
	17	17	40	57	20	95

1. Examples 18 and 19 and Comparative Examples 12 and 13

Adhesive compositions were produced in the same manner as in Example 1, except that the blending amounts of the tackifier and the softener were changed as shown in Table 5 and that 1 part by weight of AO-1, 0.5 part by weight of AO-3 and 0.5 part by weight of AO-6 were used as stabilizers. Their performance characteristics were evaluated. The results obtained are shown in Table 5.

Table 5

	Blending amount tackifier (parts by weight)	Blending amount of softener (parts by weight)	Ball tack (No.)	Adhesive strength (g/cm)	Creep resistance (min.)
Example 18	150	60	24	680	120
Example 19	70	20	17	850	250
Comparative Example 12	250	30	<3	Kraft paper was broken	30
Comparative Example 13	20	30	<3	<300	25

1 Examples 20 and 21

Adhesive compositions were produced in the same manner as in Example 1, except that the blending amounts of the tackifier and the softener were changed as shown in 5 Table 6 and that in each composition, there were used 0.5 part by weight of AO-1 as phenolic compound, 0.5 part by weight of AO-14 as sulfur-containing stabilizer, 0.5 part by weight of AO-3 as another phenolic compound, and 0.5 part by weight of AO-6 as phosphorus-containing 10 stabilizer. Their performance characteristics were evaluated. The results obtained are shown in Table.

Table 6

	Blending amount tackifier (parts by weight)	Blending amount of softener (parts by weight)	Ball tack (No.)	Adhesive strength (g/cm)	Creep resistance (min.)
Example 20	150	60	24	700	130
Example 21	70	20	17	860	255

1 Examples 22 to 24

Adhesive compositions were produced according to the recipes shown in Table 7, and their performance characteristics were evaluated. With 100 parts by weight of each block copolymer were blended 100 parts by weight of each tackifier shown in Table 7, 30 parts by weight of a softener (Sonic process Oil R-200), 0.5 part by weight of AO-1, 0.3 part by weight of AO-13, and 0.5 part by weight of AO-14.


Table 7

	Block copolymer used				Tackifier used	Ball tack (No.)	Adhesive strength (g/cm)	Creep resistance (min.)
	Polymer structure	Styrene content [S] (wt. %)	1,2-Vinyl content [V] (%)	[S]+[V]				
Example 22	BABAB	25	43	68	Aromatic-compound-modified terpene resin (Note 4)	19	710	150
Example 23	$(A-B-\frac{1}{4}Si)$ (Note 5)	25	25	50	Alicyclic saturated petroleum resin (Note 6)	17	730	120
Example 24	DABA	16	27	42	Modified aliphatic petroleum resin (Note 7)	21	760	90

- 1 Note 4: YS-resin TO-105 (mfd. by Yasuhara Yushi Co.,  
Ltd.) was used.
- Note 5: There was used a block copolymer in which about  
5% by weight of styrene was taperingly bonded to  
5 butadiene in the portion B. The number of  
tapers was 2.
- Note 6: Arkon M-90 (mfd. by Arakawa Chemical Co., Ltd.)  
was used.
- Note 7: ECR 316 (mfd. by Exxon Chemical Co., Ltd.) was  
10 used.

#### Example 25

A hot-melt adhesive composition was produced by  
blending 100 parts by weight of a block copolymer composed  
of a blend of the block copolymer used in Example 1 and  
15 20% by weight of a block copolymer of an A-B structure  
having a styrene content of 20% by weight and a 1,2-vinyl  
content of 33% with 100 parts by weight of Quintone U-185,  
30 parts by weight of Sonic Process Oil R-200, 1 part by  
weight of AO-1, 0.5 part by weight of AO-10 and 0.5 part  
20 by weight of AO-9. Performance characteristics of the  
adhesive composition were evaluated to find that it had a  
ball tack of 23, an adhesive strength of 850 g/cm, and a  
creep resistance of 170 minutes. Thus, a composition  
having a further improved initial tack could be obtained  
25 by blending the block polymer having an A-B structure.





1 Example 26

A terminally modified block copolymer having an N-methylpyrrolidone reacted residue attached to the end of polymer chain was produced by reacting N-methylpyrrolidone  
5 with a block copolymer having the same styrene content and 1,2-vinyl content as in Example 3 obtained by polymerization in cyclohexane by use of n-butyl lithium as catalyst, before deactivation. An adhesive composition was produced in the same manner as in Example 3, except  
10 that said terminally modified block copolymer was used in place of the block copolymer used in Example 3. Performance characteristics of the adhesive composition were evaluated to find that it had a ball tack of 21, an adhesive strength of 900 g/cm and a creep resistance of  
15 215 minutes. Thus, the creep resistance, in particular, was improved by the employment of the terminally modified block copolymer.

1. An adhesive composition characterized by comprising a blend of 100 parts by weight of a block copolymer (a) containing of at least one polymer block composed mainly of a vinyl aromatic hydrocarbon and at least one polymer block composed mainly of butadiene in which

(i) the content of said vinyl aromatic hydrocarbon [S] is 10 to 30% by weight,

(ii) the content of 1,2-vinyl in said butadiene portion [V] is 20 to 50%, and

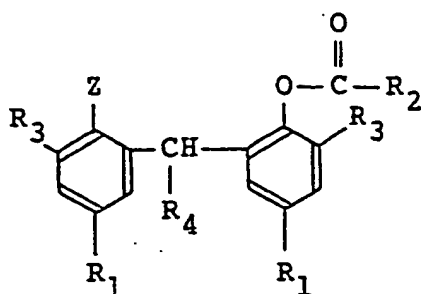
(iii) the relationship between said vinyl aromatic hydrocarbon content [S] and said 1,2-vinyl content in butadiene portion [V] satisfies the following formula:

$$40 \leq [S] + [V] \leq 70$$

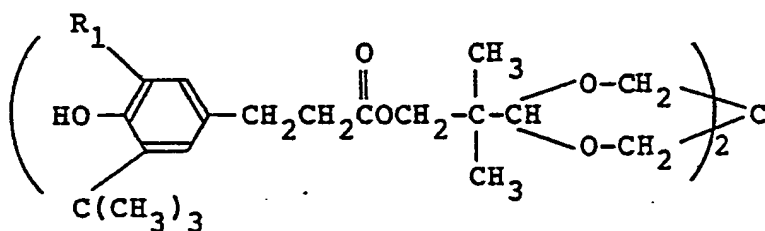
and

40 to 200 parts by weight of a tackifier (b).

2. An adhesive composition according to Claim 1, characterized by further containing, per 100 parts by weight of said block copolymer (a), 0.05 to 5 parts by weight of at least one member selected from the group consisting of phenolic compounds (c) represented by the general formula [I] or [II]:



[I], and



[II]

wherein Z is OH or  $\text{O-C(=O)-R}_2$ ,  $\text{R}_1$  is an alkyl group having 1 to 4 carbon atoms,  $\text{R}_2$  is an alkenyl group having 2 to 4 carbon atoms,  $\text{R}_3$  is a tert-butyl group or a cyclohexyl group, and  $\text{R}_4$  is a hydrogen atom or an alkyl group having 1 to 18 carbon atoms.

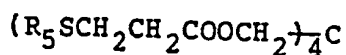
3. An adhesive composition according to Claim 2, characterized in that the phenolic compound (c) represented by the above general formula [I] is one selected from the group consisting of 2,2'-methylene-bis-(4-methyl-6-tert-butylphenol) monoacrylate, 2,2'-methylene-bis-(4,6-di-tert-butylphenol) monoacrylate, 2,2'-ethyldiene-bis-(4,6-di-tert-butylphenol) monoacrylate, 2,2'-methylene-bis-(4-methyl-6-tert-butylphenol) diacrylate, 2,2'-methylene-bis-(4,6-di-tert-butylphenol) diacrylate,

and 2,2'-ethylidene-bis-(4,6-di-tert-butylphenol) diacrylate.

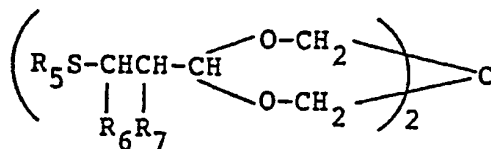
4. An adhesive composition according to Claim 2, characterized in that the phenolic compound (c) represented by the above general formula [II] is one selected from the group consisting of 3,9-bis-[2-{3-(3-tert-butyl-4-hydroxy-5-methylphenyl)propionyloxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro [5.5]undecane and 3,9-bis-[2-{3-(3-tert-butyl-4-hydroxy-5-isopropylphenyl)propinyloxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro[5.5]undecane.

5. An adhesive composition according to any of the claims 1 to 4, characterized by further containing 0.05 to 5 parts by weight of a sulfur-containing stabilizer (d) per 100 parts by weight of said block copolymer (a).

6. An adhesive composition according to Claim 5, characterized in that said sulfur-containing stabilizer (d) is at least one member selected from the group consisting of compounds represented by the general formula [III] or [IV]:



[III], and,



[IV]

wherein  $R_5$  is an alkyl group having 3 to 20 carbon atoms; and each of  $R_6$  and  $R_7$  is a hydrogen atom or an alkyl

group having 1 to 18 carbon atoms.

7. An adhesive composition according to Claim 6, characterized in that the sulfur-containing stabilizer (d) represented by the general formula [III] or [IV] is pentaerythritol-tetrakis(3-dodecylthiopropionate), or is one selected from the group consisting of 3,9-bis-(2-dodecylthioethyl)-2,4,8,10-tetraoxaspiro[5.5]undecane and 3,9-bis-(2-octadecylthiopropyl)-2,4,8,10-tetraoxaspiro[5.5]undecane.

8. An adhesive composition according to any of the claims 1 to 7, characterized by further containing 5 to 150 parts by weight of a softener (component (f)) per 100 parts by weight of said block copolymer (a).

9. An adhesive composition according to any of the claims 1 to 8, characterized in that the tackifier (b) is at least one member selected from the group consisting of terpene resin, aromatic-compound-modified terpene resins, alicyclic saturated petroleum resins, rosin esters, disproportionated rosin esters, completely hydrogenated rosin esters, aliphatic petroleum resins, and modified aliphatic petroleum resins.

10. A process for producing an adhesive, characterized by blending 100 parts by weight of a block copolymer (a) containing at least one polymer block composed mainly of a vinyl aromatic hydrocarbon and at least one polymer block composed mainly of butadiene with 40 to 200 parts by weight of a tackifier (b),

(i) said block copolymer (a) having a vinyl

aromatic hydrocarbon content [S] of 10 to 30% by weight

(ii) said block copolymer (a) having a 1,2-vinyl content of butadiene portion [V] of 20 to 50%, and

(iii) the relationship between said vinyl aromatic hydrocarbon content [S] and said 1,2-vinyl content of butadiene portion [V] satisfying the formula:

$$40 \leq [S] + [V] \leq 70.$$

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1. A process for producing an adhesive composition characterized by blending 100 parts by weight of a block copolymer (a) containing of at least one polymer block composed mainly of a vinyl aromatic hydrocarbon and at least one polymer block composed mainly of butadiene in which

(i) the content of said vinyl aromatic hydrocarbon [S] is 10 to 30% by weight,

(ii) the content of 1,2-vinyl in said butadiene portion [V] is 20 to 50%, and

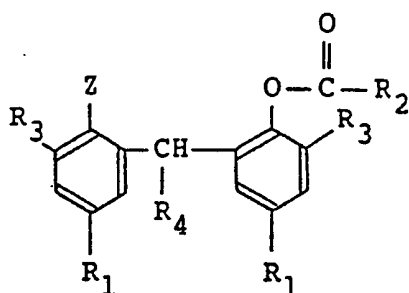
(iii) the relationship between said vinyl aromatic hydrocarbon content [S] and said 1,2-vinyl content in butadiene portion [V] satisfies the following formula:

$$40 \leq [S] + [V] \leq 70$$

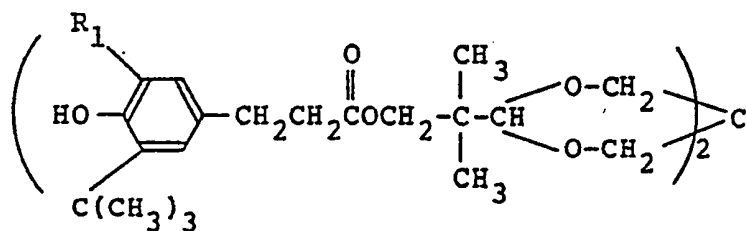
and

40 to 200 parts by weight of a tackifier (b).

2. A process according to Claim 1, characterized by further blending, per 100 parts by weight of said block copolymer (a), 0.05 to 5 parts by weight of at least one member selected from the group consisting of phenolic compounds (c) represented by the general formula [I] or [II]:



[I], and



[II]

wherein Z is OH or  $\text{O-C(=O)-R}_2$ ,  $\text{R}_1$  is an alkyl group having 1 to 4 carbon atoms,  $\text{R}_2$  is an alkenyl group having 2 to 4 carbon atoms,  $\text{R}_3$  is a tert-butyl group or a cyclohexyl group, and  $\text{R}_4$  is a hydrogen atom or an alkyl group having 1 to 18 carbon atoms.

3. A process according to Claim 2, characterized in that the phenolic compound (c) represented by the above general formula [I] is one selected from the group consisting of 2,2'-methylene-bis-(4-methyl-6-tert-butylphenol) monoacrylate, 2,2'-methylene-bis-(4,6-di-tert-butylphenol) monoacrylate, 2,2'-ethyldiene-bis-(4,6-di-tert-butylphenol) monoacrylate, 2,2'-methylene-bis-(4-methyl-6-tert-butylphenol) diacrylate, 2,2'-methylene-bis-(4,6-di-tert-butylphenol) diacrylate,



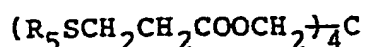
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and 2,2'-ethylidene-bis-(4,6-di-tert-butylphenol) diacrylate.

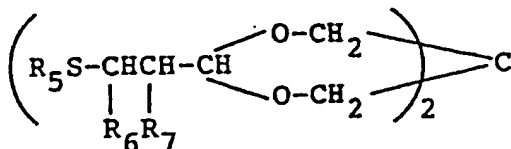
4. A process according to Claim 2, characterized in that the phenolic compound (c) represented by the above general formula [II] is one selected from the group consisting of 3,9-bis-[2-{3-(3-tert-butyl-4-hydroxy-5-methylphenyl)propionyloxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro [5.5]undecane and 3,9-bis-[2-{3-(3-tert-butyl-4-hydroxy-5-isopropylphenyl)propinyloxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro[5.5]undecane.

5. A process according to any of the Claims 1 to 4, characterized by further blending 0.05 to 5 parts by weight of a sulfur-containing stabilizer (d) per 100 parts by weight of said block copolymer (a).

6. A process according to Claim 5, characterized in that said sulfur-containing stabilizer (d) is at least one member selected from the group consisting of compounds represented by the general formula [III] or [IV]:



[III], and,



[IV]

wherein  $R_5$  is an alkyl group having 3 to 20 carbon atoms; and each of  $R_6$  and  $R_7$  is a hydrogen atom or an alkyl

group having 1 to 18 carbon atoms.

7. A process according to Claim 6, characterized in that the sulfur-containing stabilizer (d) represented by the general formula [III] or [IV] is pentaerythritol-tetrakis(3-dodecylthiopropionate), or is one selected from the group consisting of 3,9-bis-(2-dodecylthioethyl)-2,4,8,10-tetraoxaspiro[5.5]undecane and 3,9-bis-(2-octadecylthiopropyl)-2,4,8,10-tetraoxaspiro[5.5]undecane.

8. A process according to any of the Claims 1 to 7, characterized by further blending 5 to 150 parts by weight of a softener (component (f)) per 100 parts by weight of said block copolymer (a).

9. A process according to any of the Claims 1 to 8, characterized in that the tackifier (b) is at least one member selected from the group consisting of terpene resin, aromatic-compound-modified terpene resins, alicyclic saturated petroleum resins, rosin esters, disproportionated rosin esters, completely hydrogenated rosin esters, aliphatic petroleum resins, and modified aliphatic petroleum resins.

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